

Tackling master equations with a flux loop transform

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A procedure is introduced which allows to represent the dynamics of a non-equilibrium system violating detailed balance by its steady state loop fluxes as the 'states'. Surprisingly, this loop representation is found to obey detailed balance. A novel algorithm for the construction of the steady state probability densities naturally emerges, as well as a 'free energy' functional the minimization of which yields the densities and currents of the non-equilibrium steady state.

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It is one of the major open questions of physics whether there exists a general principle according to which systems far from thermal equilibrium find their quasi-stationary states. While equilibrium systems simply seek the minimum of the free energy, an analogous functional governing non-equilibrium steady states (NESS) is not known. The search for such functional has been launched in the mid-twentieth century [1, 2, 3, 4], but in spite of the many authors who have contributed to this demanding task (too many to be mentioned here, in fact), it has so far not been successful. The well-known major obstacle is the absence of detailed balance in most systems of practical relevance, i.e., the presence of non-trivial probability currents in the NESS [5]. The present paper is not meant to add yet another trial to this list, but to point out a subtle structure which is common to a wide class of non-equilibrium systems, but appears to have been overlooked so far. More specifically, a procedure is introduced which transforms a system from a representation by its configurations into a representation in the space of all possible closed loops [6] of probability flux. Surprisingly, detailed balance holds in this latter representation, providing a basis for defining a statistical potential in a quite conventional manner. This may turn out to be a useful link between equilibrium statistical physics and the dynamics of systems far from thermal equilibrium.

Consider a system which can be in any one of N configurations, \mathcal{C}_i , with probabilities p_i . The system is fully determined by the set of conditional probabilities $a_{ij}\tau > 0$ to find the system in \mathcal{C}_j at time $t + \tau$ provided it was in \mathcal{C}_i at time t , with a suitable (small) time step τ . We then have $\sum_j a_{ij} = 1 \forall i$. The probability fluxes from state i to state j are

$$\phi_{ij}(t) = a_{ij}p_i(t) \quad (1)$$

which implies that there is no memory of past transitions, i.e., the dynamics is considered a first-order Markov process. For most processes of physical interest, this can be achieved by proper definition of the states \mathcal{C}_i . The temporal evolution of the probabilities is described by a master equation,

$$\frac{dp_i}{dt} = \sum_j (p_j a_{ji} - p_i a_{ij}) \quad (2)$$

The stationary solution of eq. (2) defines the set of steady state probabilities, $P^* := \{p_i^*\}$ (the asterisk indicates steady state quantities throughout this paper).

The description outlined above is widely used for a huge range of systems, including reaction-diffusion processes [7, 8], systems biology [9, 10, 11, 12], cell migration [13], trading market dynamics [14, 15], and many other classical non-equilibrium systems of general importance [4]. For continuum systems, the Master equation is replaced by the Fokker-Planck equation, which can be obtained from eq. (2) by means of the Kramers-Moyal expansion. However, since the basic ideas of the present paper can be more clearly outlined using the discrete model, we will leave continuum descriptions to future work.

In a system with detailed balance, all currents, $j_{ij} := \phi_{ij} - \phi_{ji}$, vanish in the steady state, such that

$$p_i^* a_{ij} = p_j^* a_{ji} \quad \forall i, j \quad (3)$$

In this case, eq. (3) tells us that if one p_i^* is known, the neighboring p_j^* can be immediately obtained by multiplying p_i^* with the ratio of the corresponding transition rates, a_{ij}/a_{ji} . Continuing this procedure through the whole system, we can determine each of the probabilities, independently of the path we chose in doing so. This may be viewed as an integrability condition [16].

The steady state density can then be derived from the potential [17]

$$\mathcal{U}_i := -\ln(\Pi_{0i}/\Pi_{i0}) \quad (4)$$

where Π_{0i} is defined as the product of all rates a_{ij} traversed in going step by step from a reference state, \mathcal{C}_0 , to the state of consideration, \mathcal{C}_i . We then have the simple relation $p_i^* = \mathcal{N} \exp(-\mathcal{U}_i)$ for all i , where \mathcal{N} is to be determined from the normalization condition,

$$\sum_i p_i = 1. \quad (5)$$

However, in most systems of interest eq. (3) is not fulfilled. As a consequence, \mathcal{U}_i as determined via eq. (4) would depend upon the path chosen for its evaluation. In other words, a potential in the above sense does not exist.

For the sake of clarity, we adopt a graph theoretical representation. Consider a graph $G = (V, E)$, with vertices $v_i \in V$, each of which corresponds to a configuration of the system, \mathcal{C}_i . E is the set of directed edges, e_{ij} , connecting the vertices v_i and v_j . Each edge is associated with the corresponding rate, a_{ij} . Note that we intrinsically consider a maximal graph, in which e_{ij} exists for each pair (v_i, v_j) , although the corresponding rate constant, a_{ij} , may be zero for many edges. They may actually be thought of as arbitrarily small but finite, in order to secure ergodicity, i.e., a unique NESS. We thus have $|V| = N$, and $|E| = N^2$, since the 'tadpoles' e_{ii} are included as well.

If eq (3) is not fulfilled, what we henceforth assume, there will be finite currents in the steady state. We prove that every *stationary* current, $J^* := \{j_{ij}^*\}$, can be represented as a superposition of flux loops. By a flux loop \mathcal{L} of length s we mean a set of s vertices and s directed edges which form a closed path which is self-avoiding, i.e., no vertex is visited twice. This assures that the number of possible loops, M , is finite provided G is finite. We find $M = \sum_{s=1}^N N!/s(N-s)!$ for the number of distinct self-avoiding loops which can be formed in G , and s_k is the number of steps of the loop \mathcal{L}_k . To each loop we assign an intensity m_k^* , which means that the corresponding loop contributes a flux of strength m_k^* to *each* of the edges $e_{ij} \in E_k$. We shall now prove that there exists at least one set of numbers $\{m_k^*\}$ such that the stationary *flux* distribution, Φ^* , is equal to the superposition of loop fluxes with strengths m_k . This entails the analogous (weaker) statement for the stationary current, J^* , which is already known as the flow decomposition theorem [18].

We begin by singling out one vertex, say, v_0 . The other $N-1$ vertices are to be envisaged on a circle around it, ordered according to their index, such that all edges ending or starting at v_0 are radial directed lines. Consider now the 'triloops' $\mathcal{L}_i^3 := (\{v_0, v_i, v_{i+1}\}, \{e_{0i}, e_{i(i+1)}, e_{(i+1)0}\})$ of length $s_i = 3$. We first consider the loop $\mathcal{L}_1^3 = (\{v_0, v_1, v_2\}, \{e_{01}, e_{12}, e_{20}\})$ and set $m_1^* = \phi_{01}^*$. This fully accounts for the flux ϕ_{01}^* , but also yields a contribution of ϕ_{01}^* to the edge e_{20} . Next we consider the 'biloop' $\mathcal{L}_2^2 = (\{v_0, v_2\}, \{e_{02}, e_{20}\})$ of length $s_2 = 2$. Its strength shall be $n_2^* = \phi_{20}^* - \phi_{01}^*$, such that $\phi_{20}^* = m_1^* + n_2^*$ is fully accounted for as well. Setting now $m_2^* = \phi_{02}^* - n_2^*$ for the intensity of $\mathcal{L}_2^2 = (\{v_0, v_2, v_3\}, \{e_{02}, e_{23}, e_{30}\})$, we also account for ϕ_{02}^* , and so on. We continue in this way all around the circle, until we arrive at m_{N-1}^* . This corresponds to the last triangular loop which is left. Since Φ^* is by definition an equilibrium flux distribution, we know that $\sum_i (\phi_{0i}^* - \phi_{i0}^*)$ must vanish. Furthermore, since the contribution of each loop into v_0 vanishes as well, this balance is not affected by the loops \mathcal{L}_i^2 or \mathcal{L}_i^3 . As a consequence, since ϕ_{01}^* is already fully accounted for by m_1^* , ϕ_{10}^* must be equal to m_{N-1}^* , and $n_1^* = 0$. This shows that all net fluxes to and from v_0 can be represented by a superposition of the triloop and biloop fluxes containing v_0 .

Now we remember all intensities m_i^* and n_i^* we have

determined so far, and subtract the corresponding fluxes from Φ^* , such that there are no fluxes to or from v_0 left. All that remains are fluxes within the system $V \setminus v_0$. When subtracting the fluxes represented by the loops containing v_0 , we never violated the flux balance at any vertex, since all involved fluxes were loops, and therefore themselves balanced at each vertex. As a consequence, the field of fluxes in the truncated graph is again balanced, i.e., the sum of all fluxes to and from each vertex vanishes. We can then disregard the vertex v_0 , and proceed considering only the remaining truncated graph with $N-1$ vertices. We single out one vertex again, and make all fluxes into it vanish by subtracting triloop and biloop fluxes, as described above. Note that we will not have to update any of the intensities of the flux loops containing v_0 , since v_0 (and thus all loops containing it) do not anymore belong to the system under study.

This procedure can be repeated until a graph of just two nodes is left, which is of course a single biloop. We thus have constructed a set m_k^* such that the superposition of the corresponding loop fluxes is equal to Φ^* . This proves that every balanced flux field can be represented by a superposition of triloop and biloop fluxes. *A fortiori*, it proves that Φ^* can be represented by a superposition of a set of *any* loop fluxes, without specifying their lengths, s_k . It should be noted that we cannot assure that all loop intensities are positive. However, in all systems of relevance, the number of distinct loops, M , is much larger than the number of edges, N^2 , such that the choice of the m_k^* representing a certain Φ^* is far from unique. In many (if not in all) cases, it will be possible to exploit this freedom to choose all m_k^* non-negative [19]. We finally note that in a system with detailed balance, a trivial choice is to have only biloops, \mathcal{L}_i^2 , with strengths $m_i^* = p_i^* a_{ij} = p_j^* a_{ji}$.

Next we make use of the graph representation to obtain a pictorial idea of the dynamics of the system. In the ensemble picture, we may imagine that the NESS consists of a very large number of actors travelling on the graph G step by step, each of which represents a realization of the system. Between the steps, actors reside at the vertices, and during each step (i.e., once each time interval τ) each actor on a vertex, v_i , traverses one of its outgoing edges, e_{ij} , according to the value of the corresponding rate constant, a_{ij} . Choosing the edge e_{ii} is to stay at this vertex for another time τ . The fluxes, ϕ_{ij} , just count the total number of actors traversing the edge e_{ij} in one time step, not caring which realization they represent. Using the result obtained above, we may thus represent the the steady state fluxes by imagining that each actor is eternally orbiting a single loop, \mathcal{L}_k , with the number of actors on each loop being proportional to m_k^* . More precisely, in the NESS each loop is occupied by $q_k^* := s_k m_k^*$ actors, having exactly m_k^* actors on each of its s_k vertices.

It is clear that in reality, the realizations of the system (i.e., the actors) will choose random continuations at each step instead of orbiting the loops. In other words, there

is a random exchange of actors between loops at each step. We may imagine each actor to carry a ticket for the loop he is currently orbiting. After each step, these tickets are exchanged randomly between actors at each vertex, such that the actors are redistributed among the loops, and thus among the outgoing edges of the respective vertex. If this exchange is a microscopically balanced random process, what we will henceforth assume, it fulfills detailed balance. This is the key idea of the 'flux loop transform' to be presented.

Before we proceed, we define the functional $\chi(x, X)$, where x is an object and X is a set of objects, by

$$\chi(x, X) = \begin{cases} 1 & \text{if } x \in X \\ 0 & \text{else} \end{cases} \quad (6)$$

This allows for convenient bookkeeping of which vertex belongs to which loop. With the help of eq. (6), we can write

$$\phi_{ij}^* = \sum_k m_k^* \chi(e_{ij}, E_k) \quad \forall i, j \quad (7)$$

and

$$p_i^* = \sum_k m_k^* \chi(v_i, V_k) \quad \forall i \quad (8)$$

Together with eq. (5) this leads to the normalization

$$\sum_k s_k m_k^* = \sum_k q_k^* = 1 \quad (9)$$

By combining eqs. (7) and (8), we can express the transition rates, a_{ij} , by the loop intensities as

$$a_{ij} = \frac{\sum_k m_k^* \chi(e_{ij}, E_k)}{\sum_k m_k^* \chi(v_i, V_k)} \quad (10)$$

For now, however, we are still faced with the inverse problem, which is to determine the M numbers m_k^* from the just $N^2 + N + 1$ equations (7), (8), and (9).

This ambiguity may be greatly reduced by demanding the representation of the fluxes in the space of loops to be optimized in some sense, e.g., such as to prefer few large loops over many small ones. This can be achieved by introducing a 'penalty function', $I(\{m_k^*\}) := \sum_k g(s_k) m_k^{*\gamma}$, where $g(s) > 0$ and γ can be chosen freely. This freedom of choice reflects the fundamental impact of the observer in defining convenient coarse-grained variables for characterizing a 'self-organized' state. $I(\{m_k^*\})$ will be larger if more loops are used to represent a certain flux field. A $g(s)$ which is strongly decreasing will favor long loops, and a large exponent γ prevents too intense loop fluxes to appear. As we will see, the choice of $g(s)$ and γ will have no effect on the predictions we make on the NESS acquired by the system under study. As a particularly convenient choice, we set $g(s) = 1$ and $\gamma = 2$. One might as well employ a 1-norm, $I(\{m_k^*\}) := \sum_k g(s_k) |m_k|$, which leads to particularly few non-zero loop intensities [20, 21].

If we now require $I(\{m_k^*\})$ to be minimal under the constraints (7), (8), and (9), we directly obtain, by means of the Lagrange method,

$$m_k^* + \sum_{ij} \lambda_{ij} \chi(e_{ij}, E_k) + \sum_i \mu_i \chi(v_i, V_k) + \nu s_k = 0 \quad \forall k \quad (11)$$

where λ_{ij} , μ_i , and ν are Lagrange multipliers. Combining eqs. (7) and (8), we obtain

$$\sum_k (\chi(e_{ij}, E_k) - a_{ij} \chi(v_i, V_k)) m_k^* = 0 \quad \forall i, j \quad (12)$$

Inserting eq. (11) into eq. (12) leads to N^2 equations for the set of $N^2 + N + 1$ Lagrange multipliers. The latter are thus still under-determined, although to a lesser degree than were the m_k^* . For our choice of $\gamma = 2$, however, I just represents the distance to the origin in m^* -space. Furthermore, the above equations for the m_k^* are all linear and thus define a hyperplane. Consequently, minimization of I under the above constraints just amounts to finding the point within a hyperplane which is closest to the origin (which is unique). The freedom in the Lagrange multipliers thus defines a manifold within which the set of steady state intensities is invariant, and a solution for m_k^* is uniquely obtained from $\{a_{ij}\}$ by the procedure above.

Once $\{m_k^*\}$ is known, the quantities characterizing the NESS, P^* and the corresponding currents J^* [17], can be computed directly from eqs. (8) and (1). Since all equations required to obtain $\{m_k^*\}$ have a particularly simple structure, and only contain the rate constants, this may be seen as an alternative procedure to find the steady state solution to any master equation, once the a_{ij} are given. Its practical merits as compared to other techniques, such as inversion of the matrix of rate constants, (a_{ij}) , or the method of directed trees [17], remain to be explored. Furthermore, it should be investigated if $I(\{m_k^*\})$ can be chosen such as to guarantee a set of non-negative m_k^* . These questions will be addressed in a forthcoming paper.

Let us finally turn to the flux loop transform. Consider a graph, $H = (W, F)$, with vertices $w_k \in W$, each of which corresponds to a self-avoiding loop, \mathcal{L}_k , in G . F is the set of directed edges, f_{kl} , connecting the vertices w_k and w_l . To each vertex we assign the occupation number q_k of the corresponding loop, and each edge is associated with a transfer rate constant, b_{kl} . We shall call the operation $G \rightarrow H$ the *loop transform*. By virtue of eq. (10), its inverse exists and is unique.

To investigate the properties of the transformed graph, H , we need to specify the numbers $\{b_{kl}\}$. The probability for an actor to transfer from loop \mathcal{L}_k to loop \mathcal{L}_l , at a certain vertex v_i (of G) which is common to \mathcal{L}_k and \mathcal{L}_l ($v_i \in V_k \cap V_l$) can be directly written down considering the ticket exchange process described above. It is

$$t_{kl}^{(i)} = \frac{m_l^*}{\sum_{k'} m_{k'}^* \chi(v_i, V_{k'})} = \frac{m_l^*}{p_i^*} \quad (13)$$

Since each vertex which is common to both loops yields an independent chance to transfer from \mathcal{L}_k to \mathcal{L}_l , the total probability to do so is

$$t_{kl} = \sum_i t_{kl}^{(i)} \chi(v_i, V_k \cap V_l) = C_{kl} m_l^* \quad (14)$$

where $C_{kl} := \sum_i \chi(v_i, V_k \cap V_l) / p_i^*$. On the other hand, since the probability for an actor on loop \mathcal{L}_k to be at a certain vertex i is $1/s_k$, the rate constant for transfer from \mathcal{L}_k to \mathcal{L}_l is given by $b_{kl} = t_{kl}/s_k$. Since evidently $C_{kl} = C_{lk}$, it is clear from eq. (14) that

$$q_k^* b_{kl} = q_l^* b_{lk} \quad (15)$$

which states that there is *detailed balance* in H . We can thus apply eq. (4), replacing a_{ij} by b_{kl} , to obtain a potential \mathcal{H}_k , such that the occupation numbers q_k^* are given by

$$q_k^* = \exp(-\mathcal{H}_k) \quad (16)$$

Note that $\sum_k \exp(-\mathcal{H}_k) \equiv 1$ by normalization, eq (9).

There is still a disfigurement in using H to represent the system under study. Since H concerns only loop fluxes, all possible densities $Q = \{q_k\}$ represent flux fields Φ (in G) which are balanced at each vertex, and thus yield a time-independent density field, P . This does clearly not fulfill the master equation (2) in general, which shows that any dynamics in H , away from the steady state, has nothing to do physically with the dynamics in G as described by eq. (2). It is so far only the equilibrium state of H which has a physical meaning, namely to represent the NESS in G .

This can be amended, however, by alleviating the requirement that the q_k actors on a loop \mathcal{L}_k be evenly distributed among its vertices, as is the case in the steady state. In what follows, we thus distinguish between the occupation numbers $q_{k,i}$ at the different vertices v_i along the loop \mathcal{L}_k . The relation $\sum_i q_{k,i} = s_k m_k = q_k$ is fulfilled by definition. Furthermore, we slightly change the rule for the random redistribution of the tickets introduced above such that between any two time steps, all actors at each vertex give their tickets away, and draw new ones with a probability according to the corresponding equilibrium intensities, m_k^*/p_i^* . In the steady state, this rule yields identical results as the one before, but assures the correct dynamics away from the NESS. Any distribution $P = \{p_i\}$ can then be uniquely represented setting

$$q_{k,i} = \frac{p_i}{p_i^*} m_k^* \chi(v_i, V_k) \quad (17)$$

It should be noted that the temporal evolution of the $q_{k,i}(t)$ in H does not simply obey a Master equation analogous to eq. (2). But the dynamics of the system is, by means of the flux loop transform, presented such that all violations of detailed balance are concealed within the ('interior degrees of freedom' of the) vertices of H . As time proceeds, not only will the m_k approach their equilibrium values, but also will the occupation numbers of the different vertices corresponding to the same loop approach each other ($q_{k,i}^* = m_k^* = q_k^*/s_k$).

Remembering that the $q_k = \sum_i q_{k,i}$ approach the Boltzmann distribution (16), we see that the system as represented in H is in some sense analogous to a classical system having M discrete levels at energies $\mathcal{W}_k := \mathcal{H}_k + \ln s_k$, each of which is s_k -fold degenerate. The only difference is that in H , the $q_{k,i}$ perform a 'round-dance' within each energy level (i.e., in the vertices of H), which becomes insignificant as the steady state is reached. There is thus no physical manifestation of the violations of detailed balance in the NESS as represented in H .

Finally, we may write down an entropy in H as

$$\mathcal{S}(t) = - \sum_{k,i} q_{k,i} \ln q_{k,i} \quad (18)$$

which depends on time via the $q_{k,i}$ if the system is started away from the NESS. The latter will maximize \mathcal{S} while obeying (16). Defining thus $\mathcal{W} := \sum_k q_k \mathcal{W}_k$, it is readily checked that minimizing

$$\mathcal{F} = \mathcal{W} - \mathcal{S} = \sum_{k,i} q_{k,i} \ln \frac{q_{k,i}}{m_k^*} \quad (19)$$

which is very much reminiscent to a usual free energy functional, yields precisely the NESS, i.e., the pair $\{P^*, J^*\}$ [17]. It may thus be hoped that the loop transform outlined above opens up the possibility to apply standard equilibrium statistics formalism to a wide class of systems far from thermal equilibrium.

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